

PCTWORLD INTELLECTUAL PROPERTY ORGANIZATION
International Bureau

INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification ⁴ : C08L 101/00, 17/00, 89/06	A1	(11) International Publication Number: WO 90/00184 (43) International Publication Date: 11 January 1990 (11.01.90)
(21) International Application Number: PCT/HU89/00027 (22) International Filing Date: 16 June 1989 (16.06.89) (30) Priority data: 3371/88 29 June 1988 (29.06.88) HU (71) Applicant (for all designated States except US): DATCOOP ELEKTRONIKAI KISSZÖVETKEZET [HU/HU]; Lejtő u. 10, H-1124 Budapest (HU). (72) Inventors; and (75) Inventors/Applicants (for US only) : MAGYAR, György [HU/HU]; Budakeszi ut 55/c, H-1021 Budapest (HU). VOLK, János [HU/HU]; Szigony u. 7, H-1083 Budapest (HU). SALY, György [HU/HU]; Tusnádi u. 17/b, H-1125 Budapest (HU). HÁMORI, Péter [HU/HU]; Lejtő u. 13, H-1124 Budapest (HU). FARKAS, Antal [HU/HU]; H-2144 Kerepestarcsa (HU). TAKÁTS, Péter [HU/HU]; Diósárok u. 35, H-1125 Budapest (HU). MAGYAR, István [HU/HU]; Budakeszi ut 55/c, H-1021 Budapest (HU).		(74) Agent: PATENT AND LAW OFFICE FOR INTERNATIONAL AFFAIRS; Dalszínház u. 10, H-1061 Budapest VI (HU). (81) Designated States: AT (European patent), BE (European patent), CH (European patent), DE (European patent), FR (European patent), GB (European patent), IT (European patent), LU (European patent), NL (European patent), SE (European patent), US. Published <i>With international search report.</i>
(54) Title: POLYMER COMPOSITIONS COMPRISING WASTE RUBBER AND/OR SCRAP LEATHER AS INGREDIENT (57) Abstract The invention relates to polymer compositions containing an ingredient and workable by processing methods of thermoplastics. The essence of the composition of the invention is that it contains as ingredient 10-70 % by mass of crushed waste rubber of preferably 2-5 mm grain size and/or shredded leather fibres, as well as one or more tensides, and, if desired, additives or auxiliaries corresponding to the field of application, such as fire retardants, blowing agents, moisture-binding agents, traditional fillers, colouring agents and pigments, lubricants and stabilizers, crosslinking agents.		

FOR THE PURPOSES OF INFORMATION ONLY

Codes used to identify States party to the PCT on the front pages of pamphlets publishing international applications under the PCT.

AT	Austria	FI	Finland	ML	Mali
AU	Australia	FR	France	MR	Mauritania
BB	Barbados	GA	Gabon	MW	Malawi
BE	Belgium	GB	United Kingdom	NL	Netherlands
BF	Burkina Faso	HU	Hungary	NO	Norway
BG	Bulgaria	IT	Italy	RO	Romania
BJ	Benin	JP	Japan	SD	Sudan
BR	Brazil	KP	Democratic People's Republic of Korea	SE	Sweden
CF	Central African Republic	KR	Republic of Korea	SN	Senegal
CG	Congo	LJ	Liechtenstein	SU	Soviet Union
CH	Switzerland	LK	Sri Lanka	TD	Chad
CM	Cameroon	LJ	Luxembourg	TG	Togo
DE	Germany, Federal Republic of	MC	Monaco	US	United States of America
DK	Denmark	MG	Madagascar		
ES	Spain				

POLYMER COMPOSITIONS COMPRISING WASTE RUBBER AND/OR SCRAP
LEATHER AS INGREDIENT

Technical Field

The aim of the present invention is to provide com-
5 positions by using thermoplastic polymers (polyethylene,
polypropylene, ethylene - vinyl acetate copolymer,
poly(vinyl chloride) and its copolymers, polyurethane,
polyamide, polyester, polystyrene and styrene copolymers,
cellulose derivatives, as well as other thermoplastics),
10 which compositions are characterized in that they contain
- in addition to other modifying components - scrap
leather and/or waste rubber as ingredient in a relatively
high quantity (10-70 % by mass) as compared to the polymer
matrix and these compositions may be processed by standard
15 methods applicable for thermoplastics.

Background Art

The Austrian patent specification No. 368 446 dis-
closes a process for producing carpets made from poly-
urethane-bonded rubber granulates, wherein 10-20 parts of
20 polyurethane to be crosslinked and 100 parts of rubber
are combined mechanically (by pressing).

The Austrian patent specification No. 355 291 dis-
closes a process for producing floor coverings and carpets
by using shredded tyres; after cleaning the shredded
25 wastes are linked together under pressure with curing
materials.

The Austrian patent specification No. 339 797 des-
cribes a floor covering for sport establishments, accord-
ing to that solution 6-20 % of ground rubber of 1-10 mm
30 grain size are applied together with crushed sand or
breakstones, by using bituminous or latex binder.

The Austrian patent specification No. 370 168 dis-
closes a process for producing building elements from used
tyres and building bodies from the elements. The said des-
35 cription relates to the assembling of elements and sections

obtained from a specific dimensional cutting of tyres, as well as to the shape of the building elements.

A permeable floor covering for sporting grounds, walking surfaces and others may be known from the Austrian
5 patent specification No. 287 047. According to the concrete aim, rubber granulates of 0,1-4 mm grain size are used, combined with adhesives, e.g. with caoutchouc dispersion. The properties of the floor covering may be changed with asbestos flour, sand and cement additives and the product
10 may be dyed.

The patent specification No. DE 3 409 015 discloses a process for producing sheetlike or blocklike moulded bodies. The process comprises mixing ground rubber the grain size of which is inferior to 6 mm with prefoamed poly-
15 ethylene and crosslinking the mixture in mould, under pressure.

From the Swiss patent specification No. 601 567 a sporting ground covering has become known, the surface abrasive layer and lower damping layer of which contain
20 used tyre sections and polyurethane binder.

The patent specification No. DD 121 744 discloses a process for the utilization of used rubber granulates, especially for the purposes of road construction. The working in of the rubber granulates of about 25 mm grain size
25 obtained by grinding used tyres is carried out in concrete mixer, with cement, sand and water, thus a heat-insulating or elastic layer is formed, particularly for road construction. As binder, bitumen can also be used.

The patent specification No. DD 252 945 describes a
30 process for producing heat- and sound-insulating moulded bodies, wherein ground used tyres and phenol-formaldehyde resin binder are utilized for developing the insulating moulded bodies.

Thus the literature does not contain any data concern-
35 ing the combination of leather with polymer, and no really

up-to-date method is known for the processing and combination of waste rubber, by means of which the ingredient could be embedded in the polymer matrix.

5 The aim of the invention is to process on a large scale waste rubber, leather and plastics by developed technology and modern techniques into polymer mixtures or finished products satisfying wide-spread demands.

10 It is well-known that ingredients are mixed with different polymers in order to improve various properties or to reduce the price of the product, in many cases even at the cost of the deterioration of other, similarly important characteristics. Generally, ingredients of different type are used to improve the mechanical properties, electric features, sound- and heat-insulating capacity, resistance
15 to heat and chemicals, etc.

Disclosure of the Invention

The compositions of the invention contain waste rubber and/or scrap leather as ingredient.

20 In the polymer systems combined in melt state the rubber and leather grains duly adhere to the polymer matrix, at the same time they bring about - as elastic fillers - a surprisingly peculiar behaviour of the polymer system.

Due to their macromolecular structure, the rubber and leather ingredients are very near to the polymers from
25 physical and chemical point of view as well.

The specific surface of leather and rubber is high arising from the grinding process and owing to their fine structure, and as a result of their macromolecular nature they are in closer physical interaction with the polymer
30 matrix molecule than the traditional, small molecular ingredients of non-polymer type would be. In addition, the elastic rubber and leather ingredients of polymer type are attracted to the molecules of polymer matrix with secondary chemical forces, too.

35 As a consequence, they may be filled in the basic

material in larger quantities than the usual ingredients, without hindering the forming operations of thermoplastic polymers, besides, the characteristics of the processed product change favourably.

- 5 The properties of the compositions may be altered suitably in a wide range, primarily according as fundamentally different, fine-structure leather or rubber is used as ingredient in a wide range of concentration and with various distributions of grain size or the joint applica-
10 tion thereof is intended to reach special combinations of properties. As secondary modifying possibilities, known modifiers may be used, such as fire retardants, blowing agents, moisture-binding agents, traditional fillers, colouring agents and pigments, lubricants and stabilizers,
15 crosslinking agents, etc.

Examinations have been made with regard to the capacity of a polymer to take rubber or leather. It has proved to be capable of taking 10-70 % by mass, the physical and chemical properties of the combined polymer can be modified
20 by changing the quantity of rubber/leather and polymer, as well as by the specific surface of the filler.

After that, examinations have been carried out in such a manner that various polymers have been mixed in different degrees, whereby their properties have been added
25 up, then the above mentioned quantity of leather and/or rubber ingredient was fed. It has turned out that the physical and chemical properties of the new polymer mixtures can be changed favourably as compared to the previous ones.

During these experiments it has been noted that the
30 application of a polymer containing vinyl acetate (EVA), e.g. ESCORINE UL 0-0220 CH 1, has changed the properties of the polymer mixture in an extremely favourable manner.

It is well-known that a lot of polymers contain additives of tenside type which have lubricating effect or
35 facilitate the mixing with other polymers, as well as

- 5 -

contribute to the better formation of secondary bonds of the elastic filler (rubber/leather).

After the above conclusions, research has been carried out to the effect of how the various tensides would influence the linkage of polymer mixtures to an elastic filler. With this end in view, tensides of various types and in various quantities (0,02-5 % by mass) have been added to various quantities of polymer mixtures and to this has been added the rubber and/or leather ingredient. By means of the obtained filled polymer mixture there have been brought about materials with extremely good physical and chemical properties, changeable in a wide range and suitable for multiple utilization.

The structure of the filled polymer mixtures has been examined with electron microscope and it has been found that in case of the tenside containing filled polymer mixtures the polymers receive the filler favourably, their adhesion is very strong, the objects produced thereof tolerate deformation better, without interruption of contact.

The fillers used for polymers up to now were mainly inorganic materials, the continuity of which was broken in the polymer mixtures, therefore they resulted in unfavourable properties.

On the basis of our experiments it can be stated that the polar parts of the leather/rubber filler establish contact with the polar groups of the tenside, thus the tenside-coated filler shows outwards a nonpolar character owing to the nonpolar ends of the tenside. The filler which has become nonpolar in this way is well miscible with nonpolar polymers (e.g. polyethylene, polypropylene). As a result of the tenside, the surface is increased in which favourable contact is established between the filler and polymer mixture.

In addition to an advantageous development of the technical parameters (tensile strength, modulus of elasticity,

sound- and heat-insulating capacity, resistance to chemicals, flame-proofness, density, etc.), the practical applicability of these compositions has a great economic and environment protecting advantage.

- 5 The destruction, safe also from the point of view of environment protection, or profitable reutilization of waste rubber and leather accumulating in large quantities causes great concern all over the world. For example, scrap leather may be destroyed only in closed stoves which in-
10 volves considerable costs. The chromium containing ash arising in the stove may be stored only in poison cemeteries.

In Hungary about 13-15.000 tons of chrome-tanned scrap leather and about 40.000 tons of waste tyres arise a year.

- 15 For the compositions of the invention vegetable- and chrome-tanned scrap leathers arising in tanneries, scrap leathers of shoemaking and clothing industries, wastes of the rubber industry (first of all used tyres) may be utilized in crushed, ground state, but waste polymers are
20 also suitable (first of all poly(vinyl chloride) and polyurethane).

From the wastes fine ground material may be produced in two ways:

1. Traditional process:

- 25 Hide-fibres or ground rubbers are produced in knife mills. By this process materials of the following grain size are obtained:

	under 0,50 mm	15 %
	0,50 - 1,00 mm	35 %
30	above 1,00 mm	50 %

2. Cold grinding:

- It is a known but not wide-spread process. The waste rubbers are cooled to -80°C by standard methods, then passed through a special crushing machine with an inner temperature
35 of -75°C . As a result, fractions of the following fineness

are obtained:

under 0,50 mm	60 %
0,50 - 1,00 mm	30 %
above 1,00 mm	10 %

- 5 After the preparatory processes the obtained hide fibres and/or ground rubbers are mixed step-by-step or continuously with the polymers and additives corresponding to the individual formulae, at a temperature above the melting point of the polymer components, preferably in a mixer
10 ensuring the highest homogeneity.

The good characteristics of these compositions may be best utilized by processing on single-purpose machines, but the majority of the existing machines used for the processing of various polymers are also suitable for manu-
15 facturing these compositions economically and in good quality.

From the mixtures granulates may be produced by extrusion, from which plates, foils and other profiles may be extruded or numerous forms of products may be manufactured
20 by injection moulding and pressing.

The appropriately chosen compositions are suitable for the extrusion of 0,5-5 mm thick plates which may be further formed by vacuum forming, deep drawing, pneumatic forming.

Owing to their thermoplastic nature, the products
25 produced from these compositions can be stuff-coloured and welded with their own material. For aesthetic and functional reasons, the surface of solid or foamed products can be changed, according to the demands, with known materials (e.g. foil and sheet made of traditional polymer, fleece,
30 textile, leather, artificial leather, etc).

In certain cases the value of the product may be increased as a result of the peculiarity of the compositions of the invention that they retain their original leather and rubber smell, thereby they further the replaceability
35 of the original leather and rubber products.

The compositions of the invention enable, on the one hand, the waste leather and rubber to be processed in as large a proportion as possible and, on the other hand, the wastes arising on process and on utilization may be processed again. Thus the quantity of material intended for final destruction may be reduced to minimum, but due to their high polymer content these compositions require considerably less heating energy and they pollute the environment less than the starting scrap leather and waste rubber.

The above mentioned advantageous properties of the compositions of the invention may be utilized above all in the following fields of application:

A. Walking surfaces (mainly with fire retardant additives)

- formation of the surfaces of undergrounds, railway trains, airports and other vehicles.

B. Water insulations

- groundwater insulations
- lining of basins, flood-preventing foils
- roof boarding
- insulations for building industry.

In case of the applications under point B) there arises the advantage that the compositions may be welded on the spot with their own material.

C. Outsoles

- finished products of the shoemaking industry
- repair network of the shoemaking industry.

D. Mechanical rubber goods

- automobile carpets
- cloth-reinforced conveyors
- cloth-insertion belts
- washers, packings
- horticultural protecting foils for repeated utilization, the foils may be repaired.

The products may be reprocessed at any time, the manufacturing wastes may be returned in certain phases of the production.

Best Mode of Carrying out the Invention

The following Examples illustrate the possibilities of carrying out in practice the solution of the invention.

5 Example 1

In a laboratory inner mixer produced by Werner-Pfleiderer 30 % by mass of hide fibres, 30 % by mass of ethylene-vinyl acetate copolymer of ESCORENE UL (ESSO) 4028 type and 40 % by mass of linear poly-
10 ethylene of TIPELIN PS 380-09 type are stirred for 20 minutes at 170 °C, then the melt is cooled while pressed on a bowl into 2 mm thick plates. The cooled material is shredded with plate shears and ground in rotary grinder. From the ground material plates are
15 extruded at 180 °C on Viscosystem laboratory extruder in plate production mode. The produced plate is vacuum formable.

Example 2

Under the conditions of Example 1 ground material
20 is prepared from a mixture containing 50 % by mass of hide fibres, 25 % by mass of ethylene-vinyl acetate copolymer of ESCORENE UL 4028 type and 25 % by mass of polyethylene of TIPELIN PS 340-02 TVK type. The ground material is then processed on a BATTENFELD BSKM 30/50
25 injection moulding machine. Oblong test specimen is obtained the size of which is 65 x 50 mm and the thickness is 1 and 3 mm.

Example 3

Ground material is prepared under the conditions
30 of Example 1, with the following composition:
15 % by mass of hide fibres,
20 % by mass of ethylene-vinyl acetate copolymer of ESCORENE UL 4028 (ESSO) type,
25 % by mass of polyethylene of TIPOLEN FA 2210 TVK
35 type,

40 % by mass of aluminium hydroxide.

The melt is rolled on a bowl into 2 mm thick plates. The obtained material is favourable from the point of view of flame proofness: after having been lit and the source of
5 flame removed, it becomes self-extinguishable.

Example 4

Under the conditions of Example 1, but applying an inner temperature of 150 °C, ground material is prepared with the following composition:

- 10 30 % by mass of hide fibres,
- 30 % by mass of ethylene-vinyl acetate copolymer of ESCORENE UL 4028 (ESSO) type,
- 30 % by mass of polyethylene of TIPELIN PS 380-09 (TVK) type,
- 15 10 % by mass of azodicarbonamide of EVIPOR type (product of Egyesült Vegyiművek).

By rolling this melt into plates, a material is obtained which is foamed if put for 10 minutes in a drying oven heated to 180 °C and keeps its foam structure after cooling,
20 it has excellent heat- and sound-insulating properties.

Example 5

Under the conditions of Example 3 and with identical composition, 2 mm thick plates are rolled on a bowl in such a manner that simultaneously with charging the melt between
25 the rolls, polyethylene fleece is introduced between the melt and the roll, on one or both sides of the melt. Thus a plate is obtained to which the polyethylene fleece sticks evenly due to its partial melting.

Example 6

- 30 25 % by mass of polyethylene (TIPELIN PS 380-09, TVK),
- 25 % by mass of ethylene-vinyl acetate copolymer (ESCORENE UL 4028, ESSO) and
- 50 % by mass of ground rubber of 0,40 mm grain size are equalized. From the mixture plates are produced on an
35 extruder with slot die, at a temperature of 160-200 °C.

The obtained plate is pliable and solid.

Example 7

To the composition of Example 6 0,2-4 % of azodicarbon-
amide blowing agent is mixed and plates are prepared under
5 the conditions of Example 6. The obtained product is pliable
as rubber and has a foamed structure.

Example 8

The foamed melt produced according to Example 7 is
laminated, on calender coming after the extruder, with a
10 0,5 - 1,50 mm thick, solid polyethylene layer of TIPELIN PS
380-09 (TVK) type. One surface of the produced plate is of
foamed structure, the other is solid and smooth.

Example 9

On the extruder described in the previous examples,
15 fire-retardant plate is extruded at 180 °C which was pre-
viously converted into homogeneous mixture at 170 °C by
stirring with an inner mixer, the melt mixture was rolled
between the calender rolls into 1 mm thick plates, shredded
with plate shears, ground, then from the ground material
20 plates were extruded.

Composition of the prepared fire-retardant plate:

40 % by mass of aluminium hydroxide,
20 % by mass of polyethylene (TIPELIN FE 600-04, TVK),
20 % by mass of ethylene-vinyl acetate copolymer (ESCORENE
25 UL 4028, ESSO),
20 % by mass of ground rubber (grain size: 1,4 mm).

Example 10

Under the conditions of Example 6, plates are produced
with the following composition:

30 50 % by mass of ethylene-vinyl acetate copolymer (ESCORENE
UL 4028, ESSO),
40 % by mass of ground rubber,
10 % by mass of scrap leather made fibrous in a known way.
Owing to the combined effect of the leather and rubber in-
35 gredients, the properties of the obtained product are more

favourable than those of the plate with a composition according to Example 6.

Example 11

5 Under the conditions of Example 10 plates are manufactured, with the exception that polyethylene of TIPELIN PS 380-09 (TVK) type and ethylene-vinyl acetate copolymer of ESCORENE UL 4028 (ESSO) type are used in 1:3 ratio as polymer component.

Example 12

10 25 % by mass of polyethylene (TIPELIN PS 380-09),
25 % by mass of ethylene-vinyl acetate copolymer (ESCORENE UL 0-0220 CH 1) and
50 % by mass of ground rubber of 1 mm grain size
are equalized. From the mixture plates are produced on an
15 extruder with slot die, at a temperature of 170-200 °C. The
obtained plate is solid and vacuum-formable.

Example 13

50 % by mass of ground rubber of 1 mm grain size,
28 % by mass of polyethylene of TIPELIN PS 380-09 type,
20 2 % by mass of glycerine monostearate,
20 % by mass of polypropylene
are converted into melt on a bowl, then rolled into rubber
plates.

Example 14

25 45 % by mass of leather,
3 % by mass of zinc stearate,
22 % by mass of ESCORENE UL 0-0220 CH 1,
30 % by mass of TIPELIN FE 600-04
are mixed in a Werner-Pfeiderer inner mixer, then plates are
30 extruded therefrom at 180 °C. The obtained plate is vacuum-
-formable and dyeable.

Electron microscope tests were carried out to compare
the structure of the products of the invention and of known
products. The test results are given in Figures 1-4, wherein
35 Figure 1 shows the structure of a product containing rubber

ingredient, without tenside,

Figure 2 shows the structure of a product with identical composition, but containing tenside,

5 Figure 3 shows the structure of a product containing leather ingredient, without tenside,

Figure 4 shows the structure of a product with identical composition, containing tenside.

10 It appears from the drawings that in case of polymer compositions containing tenside, the ingredient - i.e. rubber or leather - is well embedded in the polymer matrix, in a homogeneous manner, while in case of compositions not containing tenside, agglomerated structure and considerably smaller adjoining surfaces between the ingredient and polymer matrix may be observed.

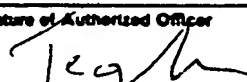
15 The surface of the products prepared from the compositions of the invention may be combined, depending on the processing technology, with natural or artificial fleece or woven textiles, with stiff materials.

C L A I M

Polymer composition containing an ingredient and workable by processing methods of thermoplastics, characterized in that it contains as ingredient 10-70 % by mass of crushed waste rubber of preferably 2-5 mm grain size and/or shredded leather fibres, as well as one or more tensides, and, if desired, additives or auxiliaries corresponding to the field of application, such as fire retardants, blowing agents, moisture-binding agents, traditional fillers, colouring agents and pigments, lubricants and stabilizers, crosslinking agents.

INTERNATIONAL SEARCH REPORT

International Application No PCT/HU 89/00027

I. CLASSIFICATION OF SUBJECT MATTER (If several classification symbols apply, indicate all) *		
According to International Patent Classification (IPC) or to both National Classification and IPC		
IPC ⁴ : C 08 L 101/00, 17/00, 89/06		
II. FIELDS SEARCHED		
Minimum Documentation Searched *		
Classification System	Classification Symbols	
Int.Cl. ⁴ :	C 08 L 101/00, 17/00, 89/06; C 08 J 11/00	
Documentation Searched other than Minimum Documentation to the Extent that such Documents are Included in the Fields Searched *		
III. DOCUMENTS CONSIDERED TO BE RELEVANT *		
Category *	Citation of Document, ¹¹ with indication, where appropriate, of the relevant passages ¹²	Relevant to Claim No. ¹³
X	US, A, 4 386 182 (J. ZIJP) 31 May 1983 (31.05.83), see totality, especially examples I-VII, comparative examples A-F.	(1)
X	GB, A, 1 246 278 (T. MIYAZATO) 15 September 1971 (15.09.71), see totality, especially examples 1,2.	(1)
X	US, A, 3 726 944 (R. BENNETT et al.) 10 April 1973 (10.04.73), see example; table I.	(1)
A	FR, A, 2 170 327 (SILLECK ENGINEERING LIMITED) 14 September 1973 (14.09.73), see examples 1,2.	(1)
A	FR, A1, 2 342 155 (MONTEDISON S.P.A.) 23 September 1977 (23.09.77), see claim 1; examples.	(1)
<div style="display: flex; justify-content: space-between;"> <div style="width: 48%;"> <p>* Special categories of cited documents: ¹⁴</p> <p>"A" document defining the general state of the art which is not considered to be of particular relevance</p> <p>"E" earlier document but published on or after the international filing date</p> <p>"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)</p> <p>"O" document referring to an oral disclosure, use, exhibition or other means</p> <p>"P" document published prior to the international filing date but later than the priority date claimed</p> </div> <div style="width: 48%;"> <p>"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention</p> <p>"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step</p> <p>"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.</p> <p>"A" document member of the same patent family</p> </div> </div>		
IV. CERTIFICATION		
Date of the Actual Completion of the International Search	Date of Mailing of this International Search Report	
06 September 1989 (06.09.89)	12 September 1989 (12.09.89)	
International Searching Authority	Signature of Authorized Officer	
AUSTRIAN PATENT OFFICE		

III. DOCUMENTS CONSIDERED TO BE RELEVANT (CONTINUED FROM THE SECOND SHEET)		
Category *	Citation of Document, with indication, where appropriate, of the relevant passages	Relevant to Claim No
A	US, A, 4 287 252 (S. DIMITER) 01 September 1981 (01.09.81).	(1)
A	GB, A, 1 113 793 (BUNZL & BIACH AKTIENGESELLSCHAFT) 15 May 1968 (15.05.68).	(1)
A	US, A, 3 489 710 (S. BONOTTO et al.) 13 January 1970 (13.01.70).	(1)

Anhang zum internationalen Recherchenbericht über die internationale Patentanmeldung Nr.

In diesem Anhang sind die Mitglieder der Patentfamilien der im obengenannten internationalen Recherchenbericht angeführten Patentedokumente angegeben. Diese Angaben dienen nur zur Unterrichtung und erfolgen ohne Gewähr.

Annex to the International Search Report on International Patent Application No. PCT/HU 89/00027

This Annex lists the patent family members relating to the patent documents cited in the above-mentioned International search report. The Austrian Patent Office is in no way liable for these particulars which are merely given for the purpose of information.

Annexe au rapport de recherche internationale relatif à la demande de brevet international n°.

La présente annexe indique les membres de la famille de brevets relatifs aux documents de brevets cités dans le rapport de recherche internationale visé ci-dessus. Les renseignements fournis sont donnés à titre indicatif et n'engagent pas la responsabilité de l'Office autrichien des brevets.

Im Recherchenbericht angeführtes Patent- dokument Patent document cited in search report Document de brevet cité dans le rapport de recherche	Datum der Veröffentlichung Publication date Date de publication	Mitglied(er) der Patentfamilie Patent family member(s) Membre(s) de la famille de brevets	Datum der Veröffentlichung Publication date Date de publication
US-A - 4386182	31-05-83	EP-A1- 23070 JP-A2-56020039 NL-A - 7905700	28-01-81 25-02-81 27-01-81
GB-A - 1246278	15-09-71	FR-A - 1587724 MY-A - 487/73 US-A - 3860539 GB-A - 1287114 US-A - 3881537 US-A - 3540306 DE-A - 1802345 DE-B2- 1802345 DE-C3- 1802345 FR-A - 1587977 GB-A - 1211167 SE-B - 330633	27-03-70 31-12-73 14-01-75 31-08-72 06-05-75 17-11-70 08-05-69 30-03-78 16-11-78 03-04-70 04-11-70 23-11-70
US-A - 3726944	10-04-73	None	
FR-A - 2170327		FR-A5- 2170327	14-09-73
see page -2-			

FR-A1- 2342155	23-09-77	AT-A - 1206/77	15-11-78
		AT-B - 350705	11-06-79
		BE-A1- 851813	25-08-77
		DE-A1- 2707980	01-09-77
		ES-A1- 456314	01-02-78
		ES-A5- 456314	25-02-78
		FR-B1- 2342155	22-02-80
		GB-A - 1513712	07-06-78
		HU-B - 178653	28-06-82
		IT-A - 1055914	11-01-82
		JP-A2-52128202	27-10-77
		NL-A - 7701836	30-08-77
		PL- P- 105438	31-10-79
		RU- P- 70499	22-06-81
		SU- D- 644396	25-01-79
		US-A - 4162996	31-07-79

US-A - 4287252	01-09-81	None
----------------	----------	------

GB-A - 1113795		None
----------------	--	------

US-A - 3489710	13-01-70	None
----------------	----------	------

For more details about this annex: see Official Journal of the European Patent Office, No. 12/82.